ELECTRONIC PROPERTIES OF METALS AND ALLOYS

Magnetoresistive oscillations in a doubly connected SFS interferometer with a ferromagnetic segment longer than the thermal coherence length

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The conductance of ferromagnetic Ni samples of macroscopic length between F/S interfaces closed by a superconductor (indium) — an SNS system in the "Andreev interferometer" (AI) geometry — is investigated. The macroscopic size of the system makes it possible to measure directly the conductance of each of the elements of the branched AI circuit and to arrange conditions under which the contribution of "side" effects, reminiscent of the proximity effect, are minimal. The $hc/2e$ oscillations of the resistance with respect to magnetic field (the Aharonov–Bohm effect in disordered conductors), with an amplitude corresponding to the positive interference contribution to the resistance of an F/S interfacial region with a thickness of the order of the coherence length of the subgap excitations upon Andreev reflection in the presence of an exchange field typical for ferromagnetic materials ($\sim$1 nm), are observed for the first time at lengths of the ferromagnetic segments exceeding the typical mesoscopic scale ($\sim 1\ \mu$m) by more than a factor of 10^3. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2747087]

I. INTRODUCTION

Together with investigations of transport coherent phenomena in mesoscopic conductors, the use of macroscopic conductors for this same purpose is of alternative interest. This is due primarily to the possibility of eliminating the ambiguity in the estimated coherence length by using samples manifestly greater than any physically reasonable coherence length for metals. Furthermore, when one is investigating a branched system, in the case of macroscopic segment lengths one can measure their conductance directly. (In mesoscopic branched systems the transport effects in the individual conductors are usually analyzed according to the behavior of the total conductance of the system, which represent a complex combination of the conductances of all its elements.[1]) Of course, a macroscopic version of the experiment will require better absolute precision of the resolution of effects of small magnitude. Furthermore, the lower relative value of the effects in macroscopic conductors requires the use of low-resistance (pure) wires.

An example of the results obtained from measurements of macroscopic samples are the results of direct four-contact measurements of the conductance of low-resistance ($\approx 10^{-8}\ \Omega$) wires of normal metals (Cu, Al, Ni) found in contact with a superconductor. The experiments were done at helium temperatures using high-resolution ($\leq 0.01\%$) potential difference measurement techniques.[2,3] We investigated the change of the resistance of the wires upon transition of the metal/superconductor (M/S) interfaces from the M/N to the M/S state. These changes were observed while the interfaces were moved apart from each other to a distance of up to 1 mm, exceeding any possible coherence lengths in metals. This meant that the observed effects pertained not to the whole length of the wire but only to a limited part of it. From the character of the response in the conductance, the effect was the direct opposite of the proximity effect. In the macroscopic experiments at the temperature transition of the interfaces to the M/S state, it was *always* manifested as an *increase* of the resistance ($R_{NS} > R_{NN}$). The corresponding contribution to the resistance of the pure wire always corresponded to a resistance of the segment of wire of length comparable to the coherence length ξ in the pure metal ($\xi \sim 1\ \mu$m). Furthermore, it was found to agree with the fundamental concepts of the theory of an increase of the elastic scattering critical of impurities upon Andreev reflection within the coherence length.[4,5] From this, in particular, it follows that in SNS systems the spatial scale of the proximity effect cannot be greater than that length. This contribution often contradicts other estimates of this scale, made from the results of indirect measurements of the conductance in size-restricted mesoscopic SNS structures (see Ref. 1).

The given example shows that resolution of an effect like the one under discussion is, generally speaking, the key to correct estimation of the spatial scales of coherence effects, including the proximity effect. Using the technique of precision measurements of small effects,[6] we investigated coherent effects in hybrid magnet/superconductor (SFS) systems with conductors of much greater than mesoscopic length. In zero external magnetic field we made direct measurements of the resistance R_F of the Ni conductors with the use of Ni/In potential interfaces set a distance of 1 mm apart.[3] As in the case of nonmagnetic conductors, at the transition of the state of the interface $F/N \rightarrow F/S$, the resistance of the ferromagnetic conductor increased every time. By comparing the value of the increase, $\delta R = R_{FS} - R_{FN}$ with the total resistance of the conductor R_{FN}, we found that it corresponds to the resistance of a layer of the ferromagnetic conductor near the F/S interface not more than a few nanometers thick. Obviously this thickness must limit the spatial scale of possible manifestation of correlation effects of the proximity type, at least in the ferromagnet under study.

The observation of the coherent effect in the SFS system settled the following question: Can effects sensitive to the phase of the order parameter in the superconductor be manifested in the conductance of ferromagnetic conductors of macroscopic size? The answer is yes.

II. EXPERIMENT AND DISCUSSION

Here we present the results of direct measurement of the conductance of Ni conductors in a doubly connected SFS configuration (in the AI geometry). Nickel samples in the geometry shown in Fig. 1 were cut by the electrospark method from a single crystal with an elastic mean free path $l_{el}>1$ μm ($RRR\approx 200$). The technology used to fabricate the system, the F/S interfaces and the In superconducting bridges, and the technique of direct measurements of the individual segments of the system were the same as in Ref. 3. The system was mounted in the end of a small lead-shielded superconducting solenoid perpendicular to the direction of the field, which was calibrated beforehand by a field probe in the end of the solenoid. The elements of the measuring circuit and the system and solenoid were secondarily shielded by a superconducting shield. Under these conditions the instability of the field depended only on the instability of the solenoid supply current I_{sol}. The absolute instability did not exceed $\delta I_{sol}=5\times 10^{-2}$ μA ($\delta H=5\times 10^{-5}$ Oe) at any value of I_{sol}. Measurements were made in the superconducting state of the In bridge for one of the samples at a temperature of 3.1 K and for the other at 3.2 K. The temperature behavior of the resistance of one of the systems, shown in Fig. 2, is typical for all the investigated systems with an In bridge.

Figures 3 and 4 show the magnetic-field oscillations of the resistance of two samples in a doubly connected S/Ni/S configuration with different aperture areas, measured for the

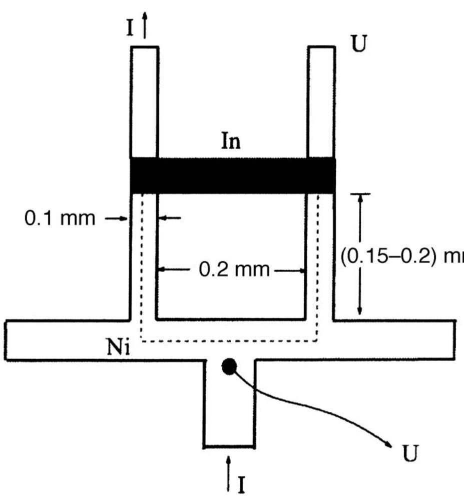

FIG. 1. Schematic diagram of the F/S system in the geometry of a doubly connected "Andreev interferometer." The ends of the single-crystal ferromagnetic (Ni) segment (dashed lines) are closed by a superconducting (In) bridge. The dimensions are indicated for the sample with the resistance oscillations shown in Fig. 3.

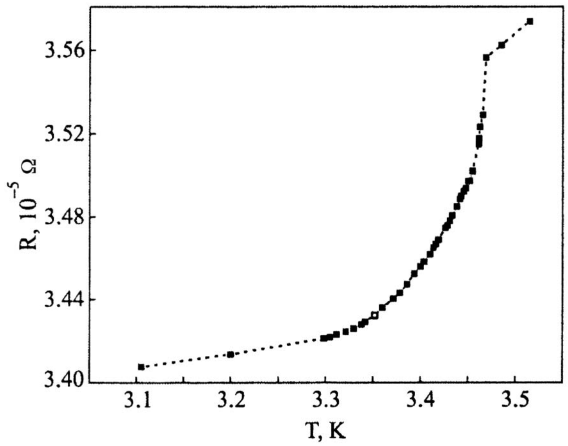

FIG. 2. Typical temperature dependence of the resistance of the In/Ni/In system in the AI geometry.

arrangement of the current and potential leads illustrated in Fig. 1. The oscillations in Fig. 3 are presented on both an absolute scale ($\Delta R_{osc}=R-R_0$, left scale) and a relative scale ($\Delta R_{osc}/R_0$, rightscale) (R_0 is the value of the resistance of the ferromagnetic segment connecting the interfaces; it is the sum of the resistances of individual parts of the segment measured independently in zero field in the absence of the bridge). Such oscillations in SFS systems in which the total length of the ferromagnetic segment (along the dashed line in Fig. 1) reaches values of the order of 1 mm have not been observed previously. We present the results for two samples and two independent measurements for opposite directions of the field with different steps in H as typical results of several measurements, confirming the reproducibility of the period of the oscillations and its dependence on the aperture area of the interferometer.

The period of the resistive oscillations shown in Fig. 1 can be seen to be $\Delta B\approx(5-7)\times 10^{-4}$ Oe. It is observed, as follows from Fig. 1, at an interferometer aperture area $A\approx 3\times 10^{-4}$ cm^2, measured from the midline of the segments and bridge. In a sample with twice the length of the sides of the interferometer and, hence, approximately twice the aperture area, the period of the oscillations turned out two be approximately half as large (solid curve in Fig. 4). For con-

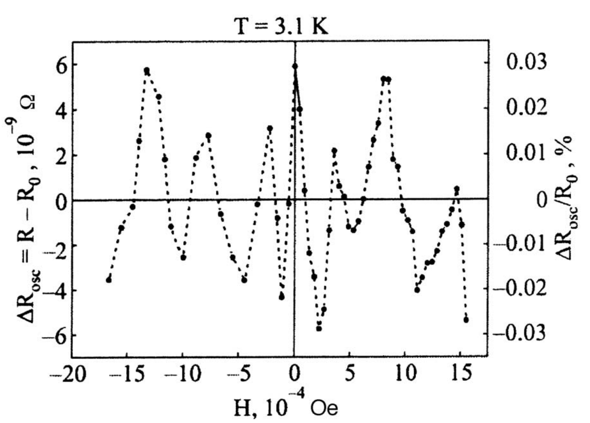

FIG. 3. The $hc/2e$ magnetic-field oscillations of the resistance of a ferromagnetic (Ni) conductor in an AI system with the dimensions given in Fig. 1, in absolute (left-hand scale) and relative (right-hand scale) units. $R_0=4.12938\times 10^{-5}$ Ω. $T=3.1$ K.

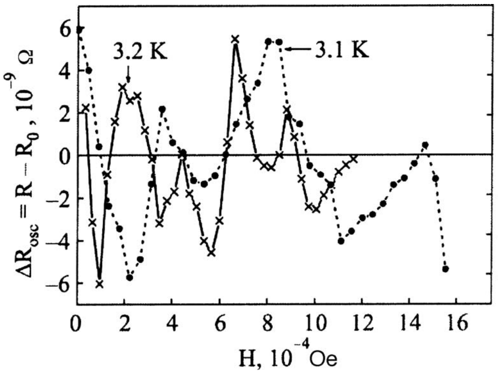

FIG. 4. The $hc/2e$ magnetic-field oscillations of the resistance of a ferromagnetic (Ni) conductor in an AI system with an aperture area twice that of the system illustrated in Fig. 1 (solid curve, right-hand scale). $R_0 = 3.09986\times10^{-6}\ \Omega$. $T=3.2$ K. The dashed curve shows the oscillations presented in Fig. 3.

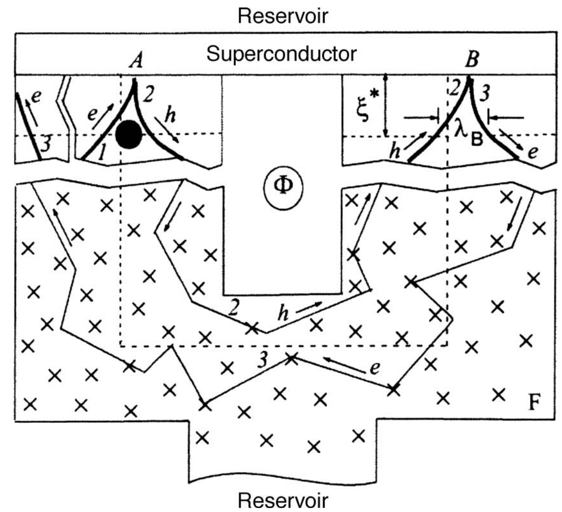

FIG. 5. Geometry of the model. See text for details.

venience of comparison the oscillations for both samples are plotted in Fig. 4. From the values of the periods of the observed oscillations it follows that, to an accuracy of 20%, the periods are proportional to a quantum of magnetic flux $\Phi_0 = hc/2e$ passing through the corresponding area $A:\Delta B \approx \Phi_0/A$. Here the relative amplitude of the oscillations for the samples $\Delta R_{\max}/\bar{R}_L \approx 0.03\%$ and 0.01%, which corresponds to a relative value of the coherent effects measured independently in a configuration with open interfaces (see also Ref. 3).

Obviously, the dependence of the conductance of the system on the magnetic flux means, in particular, that the conductance depends on the *total* length of the contour enclosing the magnetic flux. In our case this length actually corresponds to the macroscopic distances L (the length of the dashed line in Fig. 1) between the Ni/S interfaces ($L \approx 0.7$ mm and 1.1 mm, respectively, for the first and second samples). But oscillatory behavior of the conductance is possible if the phases of the electron wave functions are sensitive to the phase difference of the order parameter in the superconductor at the interfaces.[7] Consequently, the latter should be related to the diffusion trajectories of the electrons on which the "phase memory" is preserved within the whole length L of the ferromagnetic segment. This means that the oscillations are observed in the regimes $L \sim L_\varphi = \sqrt{D\tau_\varphi} \gg \xi_T = \sqrt{\hbar D/T}$ (D is the diffusion coefficient, ξ_T is the coherence length of the metal, over which the proximity effect vanishes, and τ_φ is the dephasing time). It is well known that the possibility for the Aharonov–Bohm effect to be manifested under these conditions was proved by Spivak and Khmel'nitskiĭ,[7] although the large value $L_\varphi \sim L$ coming out of our experiments is somewhat unexpected. It must mean that the destructive contribution of the inelastic scattering in the ferromagnet (as in normal metals[2]) can already be extremely small at not very low helium temperatures. In this regard it makes sense to make a qualitative estimate of the possible scale of L_φ for that temperature region. The value of the temperature contribution δR_T to the resistance in this region in comparison with the residual resistance R_0 for metals with $l_{\mathrm{el}} > 10\ \mu$m ($D \sim 10^5$ cm^2/s) is usually of the order of $\delta R_T/R_0 \sim 10^{-3}$–$10^{-4}$, and the electron–phonon relaxation time

$$\tau_{e-\mathrm{ph}} \sim \frac{l_{\mathrm{el}}}{v_F}\frac{1}{\delta R_T/R_0} \sim (10^{-7} - 10^{-8})\mathrm{s},$$

while the electron–electron relaxation time $\tau_{e-e} \sim 10^{-8}$ s. Hence the possible dephasing length at helium temperatures is $L_\varphi = \sqrt{D\tau_\varphi} \sim 0.3$–$1$ mm.

Under these conditions the nature of the observed oscillations, according to the arguments made by Spivak and Khmel'nitskiĭ in a discussion of electron diffusion in a simply connected SNS system,[7] is as follows. According to those arguments, in a metal, regardless of the sample geometry (the parameters $L_{x,y,z}$), there always exists a finite probability of constructively interfering transport trajectories, the oscillatory contribution of which does not average out. Such trajectories coexist with destructively interfering ones, the contributions of which average practically to zero.[7,8] These features were manifested, e.g., in the experiment of Sharvin and Sharvin,[9] in which the constructive interference was observed in a magnesium cylinder with a wall thickness of at least $100\lambda_B$ (λ_B is the quasiclassical thickness of a trajectory). Under the condition $L_{x,y,z} \leq L_\varphi$, only the number (probabilities) of constructively and destructively interfering trajectories depends on the values of the parameters $L_{x,y,z}$. The doubly connected geometry "improves" this relation some more, artificially organizing the doubly connected trajectories capable of constructive interference. As a result, the probability of the appearance of trajectories capable of interfering constructively increases. The difference between the doubly connected SNS and SFS systems consists in the fact that in the presence of a superconducting segment the ends of the electron (e) and hole (h) diffusion trajectories, coupled by Andreev reflection, on each of the S interfaces can coincide in the first case and, as is shown below, cannot coincide in the second case.

Figure 5 shows typical trajectories arising in a doubly connected SFS system upon Andreev reflection. Because of the Larmor curvature under the influence of the exchange field of the ferromagnet, the trajectories e (*1*), h (*2*) [h (*2*), e (*3*), etc.] diverge to the trajectory thickness λ_B (Ref. 3), losing coherence over a distance $\xi^* = \sqrt{2\lambda_B r_{\mathrm{exch}}}$ from the interface[10] (r_{exch} is the Larmor radius in the exchange field

$H_{\rm exch} \approx k_B T_c/\mu_B$ (for Ni $r_{\rm exch} \sim 1$ μm); T_c, k_B, and μ_B are the Curie temperature, Boltzmann's constant, and the Bohr magneton, respectively). The phase shifts acquired by (for example) an electron (*3*) and hole (*2*) on the trajectories connecting the interfaces are equal, respectively, to[11]

$$\phi_e = (k_F + \varepsilon_T/\hbar v_F)L_e + 2\pi\Phi/\Phi_0 = \phi_{0e} + 2\pi\Phi/\Phi_0,$$

$$\phi_h = -(k_F - \varepsilon_T/\hbar v_F)L_h + 2\pi\Phi/\Phi_0 = \phi_{0h} + 2\pi\Phi/\Phi_0. \quad (1)$$

Here ε_T, k_F, and v_F are the energy, measured from the Fermi level, the modulus of the Fermi wave vector, and the Fermi velocity, respectively.

Since the trajectories of an $e-h$ pair are spatially incoherent, their oscillatory contributions, proportional to the squares of the probability amplitudes, should combine additively:

$$|f_{h(2)}|^2|f_{e(3)}|^2 \sim \cos\phi_e + \cos\phi_h \sim \cos(\phi_0 + 2\pi\Phi/\Phi_0), \quad (2)$$

where ϕ_0 is the relative phase shift of the independent oscillations, equal to

$$\phi_0 = \frac{1}{2}(\phi_{0e} + \phi_{0h}) = \frac{\varepsilon_T}{\varepsilon_L}\frac{L_e + L_h}{2L}, \quad (3)$$

where $\varepsilon_L = \hbar v_F/L$, $\varepsilon_T = k_B T = \hbar D/\xi_T^2$. Hence it follows that any spatially separated e and h diffusion trajectories with $\phi_0 = 2\pi N$, where N is an integer, can be phase coherent. Clearly this requirement can be satisfied only by those trajectories whose midlines along the length coincide with the shortest distance L connecting the interfaces. In this case $(L_e + L_h)/2L$ is an integer, since $L_{i(e,h)}, L \sim l_{\rm el}$ and $(L_{i(e,h)}/L) = m(1+\alpha)$, where $\alpha \ll 1$. Furthermore, $(\varepsilon_T/\varepsilon_L)/2\pi$ is also an integer n to an accuracy of $n(1+\gamma)$, where $\gamma \approx d/L \ll 1$ (d is the transverse size of the interface). In sum, for all the transitions considered

$$\cos(\phi_0 + 2\pi\Phi/\Phi_0) \sim \cos(2\pi\Phi/\Phi_0). \quad (4)$$

This means that the contributions oscillatory in magnetic field of all the trajectories should have the same period. Taking into consideration the quasiclassical thickness of a trajectory, we find that the number of constructively interfering trajectories with different projections on the quantization area, those that must be taken into account, is of the order of $l_{\rm el}/\lambda_B$. However, over the greater part of their length, except for the region ξ^*, where $l_{\rm el}/\lambda_B$, the trajectories are spatially incoherent. They lie with equal probability along the perimeter of the cross section of a tube of radius $l_{\rm el}$ and axis L, and therefore outside the region ξ^* they average out. Constructive interference of particles on these trajectories can be manifested only over the thickness of the segment ξ^*, reckoned from the interface, where the particles of the $e-h$ pairs are both phase- and spatially coherent. In this region the interaction of pairs with an impurity, as mentioned in the Introduction, leads to a resistive contribution. When the total length of the trajectories is taken into account, the value of this contribution for one pair should be of the order of ξ^*/L. Accordingly, one can expect that the amplitude of the constructive oscillations will have a relative value of the order of

$$\delta R^{\xi^*}/R_L \approx \frac{\xi^*}{L}\frac{l_{\rm el}}{l_{\rm el}^{\xi^*}} \sim l_{\rm el}^L/L, \quad (5)$$

($l_{\rm el}^{\xi^*} \sim \lambda_B$; Ref. 3), i.e., the same as the value of the effect measured with the superconducting bridge open. Our experiment confirms this completely: for the samples with the oscillations shown in Figs. 3 and 4, $\delta R^{\xi^*}/R_L \approx 0.03\%$ and 0.01%, respectively. This is much larger than the total contribution of the destructive trajectories, which in the weak-localization approximation are of order $(\lambda_B/l_{\rm el})^2$ and can lead to an increase of the conductance.[8] One should also note that the property (4) of the oscillations under discussion presupposes that the resistance for $H=0$ will decrease as the field is first introduced, and this, as can be seen in Figs. 3 and 4, agrees with experiment.

We note that in mesoscopic samples, as a rule, $l_{\rm el}^{\xi^*} \leq \xi^*$. In this case the contribution (5) can be comparable with the weak-localization correction. For example, in Ref. 12, where an SFS system with a mesoscopic ferromagnetic segment of length ~ 0.1 μm was investigated, the relative amplitude of the oscillations was of the order of 10^{-4}. If this is compared with the resistance of the whole segment in accordance with the rules of circuit theory, one obtains the same estimate as ours for the upper limit of the spatial scale for the proximity effect in the ferromagnets, which does not exceed the usual (singlet) scale for this effect (~ 1 nm).

III. CONCLUSION

We have investigated the conductance of a sample of ferromagnetic Ni of macroscopic length between F/S interfaces closed by a superconductor (indium) at helium temperatures. The configuration of the SFS system corresponded to the "Andreev interferometer" geometry. We have for the first time observed the $hc/2e$ oscillations of the resistance in magnetic field in an AI with a ferromagnet segment of more than 10^3 times greater length than in mesoscopic structures. A physical explanation is offered for the parameters of the oscillations observed. We have found that the amplitude of the oscillations corresponds to the value of the positive resistive contribution to the resistance from a ferromagnetic layer several nanometers thick adjacent to the F/S interface. We have demonstrated that the scale of the proximity effect cannot exceed that thickness. The oscillations observed in a disordered conductor of an SFS system of the order of 1 mm in length (the solid-state analog of the Aharonov–Bohm effect[13]) attests to a macroscopic scale of the diffusion dephasing length in sufficiently pure metals, including ferromagnetic ones, even at not too low helium temperatures.

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